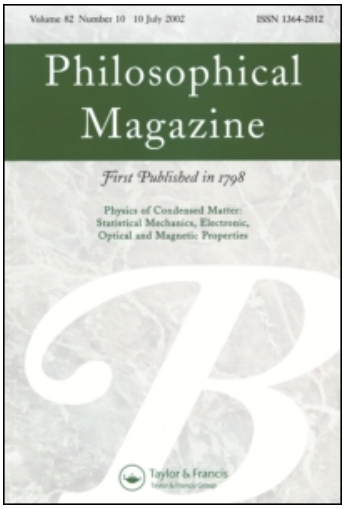

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Domain growth on percolation structures

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ABSTRACT

We discuss the dynamics of phase transformations following a quench from a high-temperature disordered state to a state below the critical temperature in the case in which the system is not translationally invariant. In particular, we consider the ordering dynamics for deterministic fractal substrates and for percolation networks by means of two models and for both a non-conserved order parameter and a conserved order parameter. The first model of phase separation employed contains a spherical constraint which enables us to obtain analytical results for Sierpinski gaskets of arbitrary dimensionality and Sierpinski carpets. The domain size evolves with time as $R(t) \sim t^{1/d_w}$ in the non-conserved case and as $R(t) \sim t^{1/2d_w}$ in the conserved case. Instead, the height of the peak of the structure factor increases as $t^{d_s/2}$ and $t^{s_s/4}$ respectively. These exponents are related to the random walk exponent d_w and to the spectral dimension d_s of the Laplace operator on the fractal lattice

The second model studied is generated from a standard Ginzburg–Landau free-energy functional on a Sierpinski carpet and random percolation structures above the percolation threshold. We consider the growth laws for the domain size $R(t)$ and the droplet size distribution.

§ 1. INTRODUCTION

The comprehension of the kinetics of phase ordering has undergone a great progress in the last decade owing to the use of analytical solutions of idealized models, numerical techniques and new theoretical ideas. Among these the concept of dynamical scaling has played an important role. The purpose of this work is to examine the late time scaling regime of some models of phase separation on various fractal supports with particular attention to the dynamic behaviour, which follows a quench from a high-temperature state to a low-temperature final state (Binder 1991, Bray 1994).

An example of a process of this kind is provided by a binary fluid mixture initially in equilibrium at temperature $T_i > T_c$, that is above the critical consolute temperature. If the temperature is lowered below T_c, the new equilibrium states correspond to two immiscible phases, characterised by different compositions. However, equilibrium is not achieved instantaneously; on the contrary the relaxation time diverges with the size of the system. After an initial stage, the system displays a typical domain structure. The average domain size $R(t)$ grows in time with a characteristic power law

$$R(t) = t^{1/z},$$

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where t is the time and z the growth exponent. The scaling hypothesis asserts that in the late stage $S(k, t) \equiv \langle \phi(\mathbf{k}, t)\phi(-\mathbf{k}, t)\rangle$ has the form

$$S(k, t) \sim R^d(t) g(kR),$$

where d is the space dimension, k is a wave-vector and $g(x)$ is a scaling function.

In spite of the fact that random or highly irregular geometries are ubiquitous in nature, when constructing physical models one usually assumes periodic lattices or regular spaces. The latter systems are much easier to study than those whose properties vary from one point to the other, but they cannot exhaust the possibilities provided by observation of the real world.

As Bravais lattices correspond to the need of describing perfectly periodic crystals, fractals have been invented in order to deal with systems which lack such regularity but still possess scale invariance under dilation or self-similarity. Among the most common physical systems of this kind are some porous materials, fractures and percolation clusters, just to mention a few. Amorphous and disordered materials are also often intimately related to fractals. In fact, one can model their geometrical structure by means of deterministic and stochastic self-similar objects. From the theoretical point of view these lattices are a geometric realization of non-integer dimensionality appearing in the ϵ expansion method.

The present paper is organized as follows: in § 2 we introduce the two dynamical models employed, referred to as case (*a*) and case (*b*); the first admits analytical solutions and represents a useful benchmark, whereas the second provides a more realistic description of the phase separation process. In § 3 we report the method of solution and the main results of model (*a*) for both conserved order parameter (COP) and non-conserved order parameter (NCOP) dynamics on Sierpinski gaskets (SGs) of arbitrary embedded dimension. In § 4 we consider the ordering dynamics of a deterministic Sierpinski carpet using model (*b*). Finally in § 5 we discuss the dynamics of model (*b*) for a random percolation network, above the percolation threshold.

§ 2. Models

In the present paper we shall investigate the ordering kinetics of some scalar field models for fractal lattices of varying Haussdorff dimensions d_f and for stochastic percolation lattices. The deterministic structure that we have considered are SGs of arbitrary embedding dimensions d, the Toblerone lattice in $d = 3$ and the Sierpinski carpet in $d = 2$.

To begin with we shall define at each lattice site a soft spin variable ϕ_i, where i is the site label and introduce a free-energy functional which describes the interaction of the field at a given site with its surroundings.

Following the Ginzburg–Landau (GL) approach we consider the evolution of the order parameter ϕ_i after a quench at temperature T_f to be described by the equation.

$$\frac{\partial \phi_i(t)}{\partial t} = -M_{il} \frac{\delta F[\{\phi_i\}]}{\delta \phi_l} + \eta_i(t)$$

$$= -M_{il}[\Delta_{lj}\phi_j(t) + f'(\phi_l)] + \eta_i(t), \tag{1}$$

where summation over repeated indices is assumed. The function $f(\phi)$ represents the local part of the free-energy density and $f'(\phi_i)$ stands for its first derivative with

respect to ϕ_i. The form of $f(\phi)$ is rather arbitrary but must have a double-well structure in order to account for phase separation. In the present paper we shall consider two possible choices for $f(\phi)$, namely model (*a*) which is a model with local nonlinearity given by

$$f = \sum_i^N \left(-\frac{r}{2}\phi_i^2 + \frac{g}{4}\phi_i^4 \right) \tag{2}$$

or model (*b*) which is a model in which the nonlinear term is non-local and given as a global character (Marini Bettolo Marconi *et al.* 1997) by

$$f = \sum_i^N \left[-\frac{r}{2}\phi_i^2 + \frac{g}{4N}\left(\sum_i^N \phi_i^2 \right)^2 \right]. \tag{3}$$

The motivation for the latter choice is that both the relevant equilibrium and the dynamical properties can be solved, thus providing useful insights into the general aspects of the physics of growth processes on non-periodic structures. This model, which is equivalent to the vector O($\mathcal{N}$) model in the large-$\mathcal{N}$ limit, or to the spherical model of Berlin and Kac (1952).

This kind of constraint is much softer than eqn. (2) since it does not act on each site, but globally over the whole volume. In the following we shall denote it as a *globally constrained model* or *spherical model* (Coniglio and Zannetti 1990) in contrast with the model in eqn. (2) where the constraint is local.

The price that one pays by choosing model (*a*) is the loss of sharp interfaces between two coexisting phases. As Abraham and Robert (1980) showed several years ago, the spherical model in a zero external field displays two ordered phases below the critical temperature, but no phase separation. Equivalently one can say that a planar interface between two coexisting phases is unstable, owing to the presence of long-wavelength excitations analogous to spin waves, an instability much stronger than that due to the presence of capillary waves in the scalar order parameter case (Marini Bettolo Marconi and Gyorffy 1989†, 1990). As a consequence, while this choice is very convenient for analytic calculations, it changes the structures of the non-uniform solutions in the static limit. Nevertheless, in spite of this fact, the model has a rich phenomenology as we shall see below and the approach to equilibrium remains highly non trivial.

In both cases, one assumes that $r < 0$ and $g > 0$, because it accounts for phase separation.

The matrix $\mathbf{\Delta}$ is the discrete version of the Laplacian operator defined on the structure under scrutiny, while $\mathbf{M}$ is a kinetic operator taking the form $\Gamma\delta_{il}$ for the NCOP and $-\Gamma\Delta_{il}$ for the COP, where Γ is a kinetic coefficient. Through the diffusive term $\mathbf{\Delta}$ a given cell is coupled to the nearest neighbours whose number Z may in principle vary from site to site. Since we consider only connected structure $Z \geqslant 1$, the noise $\eta_i(t)$ is assumed to be Gaussian with zero average and variance satisfying the fluctuation–dissipation relation $\langle \eta_i(t)\eta_j(t') \rangle = 2M_{il}T_f\delta(t-t')$, where T_f is the temperature of the final state.

† These workers have estimated the spectral dimension of the three-dimensional Sierpinski sponge to be $d_s = 2{\cdot}89$; thus the spherical model should have a real transition at finite temperatures.

§3. Soluble models

In this section we shall briefly review the static and dynamical properties of the spherical version of GL model (Martini Bettolo Marconi and Petri 1997a,b). The important feature which leads us to consider the spherical model was the possibility of obtaining in closed form the solution once the spectrum of the eigenvalues of the Laplace operator was known.

Without loss of generality, we start from the eigenvalue problem

$$-\Delta_{ij} u_j^\alpha = \epsilon_\alpha u_i^\alpha,$$

where the u_i^α are the eigenvalues associated with $\mathbf{\Delta}$ and ϵ_α are the eigenvalues.

After expanding the field ϕ as a linear superposition of modes according to

$$\phi_i(t) = \sum_{\alpha=0}^{N-1} \tilde{\phi}_\alpha(t) u_i^q.$$

we obtain within the large-N limit (Marini Bettolo Marconi and Petri 1997b) the following equation for the equal-time correlation:

$$C(\epsilon_\alpha, t) = \langle |\tilde{\phi}_\alpha(t)|^2 \rangle,$$

$$\frac{\mathrm{d}}{\mathrm{d}t} C(\epsilon_\alpha, t) = -2\epsilon_\alpha^p \Gamma[\epsilon_\alpha + r + gS(t)] C(\epsilon_\alpha, t) + 2\Gamma \epsilon_\alpha^p T_\mathrm{f}, \tag{4}$$

where $p = 0$ for the NCOP and $p = 1$ for the COP and we have defined

$$S(t) \equiv \frac{1}{N} \sum_{\alpha=0}^{N-1} C(\epsilon_\alpha, t) = \frac{1}{N} \sum_{i=1}^{N} \langle \phi_i^2(t) \rangle. \tag{5}$$

By the symbol $\langle \, . \, \rangle$ we denote averages over initial conditions and thermal histories.

The static equilibrium properties are found from eqn. (4) by determining self-consistently

$$\lim_{t \to \infty} [S(t)] = S_\infty = \frac{T_\mathrm{f}}{N} \sum_{\alpha=0}^{N-1} \frac{1}{\epsilon_\alpha + r + gS_\infty}. \tag{6}$$

Within the spherical model the vanishing of the quantity $r + gS_\infty$ as $N \to \infty$ signals the appearance of the low-temperature ordered phase below a critical temperature $T_\mathrm{c} = -r/gB(0)$, where (Coniglio and Zannetti 1990)

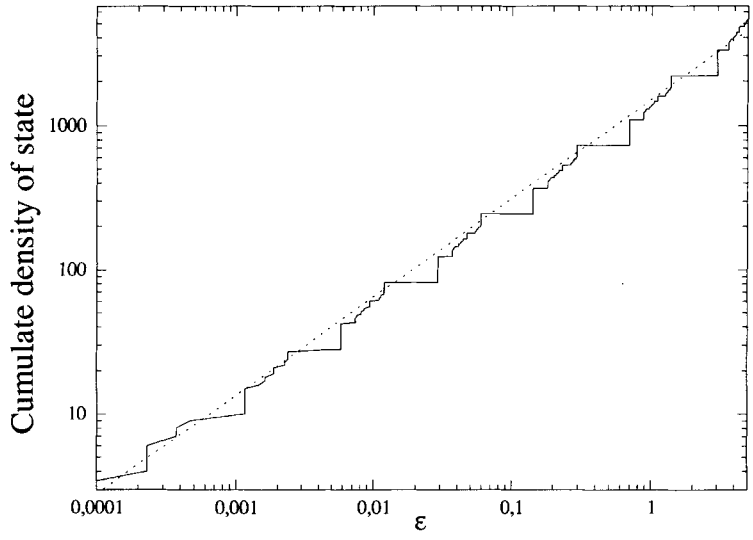
$$B(0) = \frac{1}{N} \sum_{\alpha=1}^{N-1} \epsilon_\alpha^{-1}.$$

In turn, the existence of a non-vanishing critical temperature implies the finiteness of $B(0)$ as $N \to \infty$.

The program for computing the spectrum of $\mathbf{\Delta}$ can be implemented for a finite lattice numerically, but in some cases it can even be obtained in an analytical form. Besides the spectra relative to Bravais lattices and quasicrystals in the literature we find the spectrum of $\mathbf{\Delta}$ on SGs for general embedding dimension d and fractal dimension $d_\mathrm{f} = \ln(d+1)/\ln 2$.

Its eigenvalues are contained in the interval $\epsilon_\alpha \in [0, d+3]$ and their integrated density of states is shown in fig. 1.

Fig. 1



Integrated density of states against smoothed density of states approximation for a SG with $d = 2$. $N(c) \approx c^{d_s/2}$ with $D_s = 2\ln 5/\ln 3$.

On the SG we have obtained the behaviour of B(0) in two different ways: first we observe the sum picks out its largest contribution from the smallest elements of the spectrum, which can be approximated for n sufficiently large by

$$\epsilon_\alpha \sim E_0 \frac{1}{(d+3)^{(n-\alpha)}} = E_0 \left(\frac{2^\alpha}{2^n}\right)^{d_w}, \tag{7}$$

where E_0 is a constant. In eqn. (7) we have defined the exponent

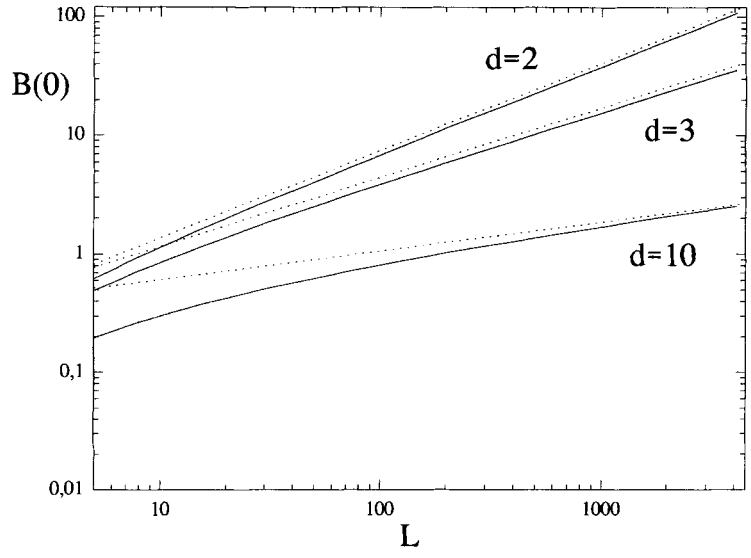
$$d_w = \frac{\ln(d+3)}{\ln 2},$$

the results of which coincide with the random walk fractal dimension on the SG (Rammal and Toulouse 1983, Rammal 1984). One the other hand the degeneracy of the generic eigenvalue ϵ_α is proportional to $(d+1)^\alpha = 2^{\alpha d_f}$. Therefore $B(0)$, which gives the inverse temperature of the model, diverges as $L^{d_w - d_f}$, where $L = N^{1/d_f}$ is the linear size of the system in lattice units, since $d_s = 2d_f/d_w < 2$ for arbitrary embedding dimension d.

In the thermodynamic limit the sum over discrete eigenvalues can be converted into the integral $B(0) = \int_{\epsilon_{min}} d\epsilon\, \rho(\epsilon)/\epsilon$, where we have employed an approximation for the density of low-frequency states $\rho(\epsilon) \sim \epsilon^{d_s/2-1}$ (fig. 2) and noticed that for large systems the smallest positive eigenvalue $\epsilon_{min} \sim E_0 L^{-d_w}$, leading to the result $B(0) \sim \epsilon_{min}^{d_s/2-1} \sim L^{d_w - d_f}$ for $d_s < 2$.

Thus, within the spherical model, T_c is lowered to zero and the ordered phase shrinks along the line $T_f = 0$, for all SGs of arbitrary embedding d; the existence of a finite-temperature phase transition requires a spectral dimension larger than two in agreement with the work of Gefen *et al.* (1984) and Cassi (1992). This can be achieved if the ramification number is infinite. The absence of a finite-temperature

Fig. 2



Double-logarithmic plot of the function $B(0)$ against the linear size L for different values of the embedding dimension d($d = 2$, $d = 3$ and $d = 10$): (- - - - -), behaviour $L^{d_w - d_f}$ for each case.

phase transition is a consequence of a generalized Mermin (1967) theorem, which states that a continuous symmetry cannot be spontaneously broken for $d_s \leqslant 2$.

Although the dynamic properties in the NCOP case can be determined exactly for arbitrary time t, we shall consider only the late scaling regime. Since eqn. (4) is formally linear, it can be integrated yielding, for $T_f = 0$,

$$C(\epsilon_\alpha, t) = C(\epsilon_\alpha, 0) \exp\{-2\Gamma[\epsilon_\alpha t + Q(t)]\}. \tag{8}$$

The auxiliary function $Q(t) = \int_0^t dt'[r + gS(t')]$, requires in order to derive the scaling behaviour of $C(\epsilon_\alpha, t)$ is determined by

$$\frac{dQ(t)}{dt} = r + gJ_d(t) \exp[-2\Gamma Q(t)], \tag{9}$$

where $J_d(t)$ for uncorrelated initial conditions $C(\epsilon_\alpha, 0) = C_0$ is given by

$$\begin{aligned} J_d(t) &= \frac{1}{N}\sum_\alpha C_0 \exp(-2\Gamma\epsilon_\alpha t) \\ &\sim C_0 \int d\epsilon\, \rho(\epsilon) \exp(-2\Gamma\epsilon t) \\ &\sim C_0 K_d t^{-d_s/2} \end{aligned} \tag{10}$$

where K_d is a dimensionality dependent constant. To proceed analytically we have employed the scaling Ansatz for $\rho(\epsilon)$. Asymptotically we find that the correlation function assumes the form

$$C(\epsilon_\alpha, t) \sim C_0 t^{d_s/2} \exp(-2\Gamma\epsilon_\alpha t). \tag{11}$$

Note that the height of the peak of eqn. (11) represents the zero amplitude of the homogeneous fluctuation and grows in time as $t^{d_s/2}$. Using eqn. (7) we can rewrite eqn. (11) in the scaling form

$$C(\epsilon_\alpha, t) \sim C_0 t^{d_s/2} \exp(-2\Gamma E_0 q^{d_w} t), \tag{12}$$

where we have introduced the quantity $q = 2^\alpha/2^n = m/L$ in order to stress the striking analogy with the standard lattice case, where a similar formula holds after replacing $d_s \to d$ and $d_w \to 2$.

Such a relation should be compared with the scaling function pertaining to standard lattices, where $D_s \to d$ and $d_w \to 2$, and the structure factor displays the scaling behaviour $C(q, t) \sim t^{d/2} \exp(-q^2 t)$.

We remark that the quantity q plays the role of the inverse of the wavelength, in agreement with the Alexander–Orbach (1982) definition of a characteristic length on fractals. As we have already observed during the study of the equilibrium properties of the system, there is no genuine phase transition at finite temperature owing to the existence of large-amplitude low-energy modes, the analogue for these lattices of long-wavelength fluctuations, which destroy the long-range order (Goldstone modes) on Euclidean lattices.

In parallel with the standard case we identify the pre-factor $t^{d_s/2}$ with R^{d_f} and deduce that $R(t) \sim t^{1/d_w}$, which represents the NCOP evolution law on fractal supports. Since $d_w > 2$ there is a slowing down of the growth caused by the delay of the diffusing particles in the fractal structure.

To summarize, the NCOP agrees with the ordinary scaling hypothesis that the typical domain size $R(t)$ is the only relevant length during the growth and the structure function has the form $C(\epsilon, t) \sim R(t)^{d_f} F[\epsilon^{1/d_w} R(t)]$, with $F[x]$ a universal time-independent shape function.

The dynamics for the COP case is described by the structure function

$$C(\epsilon_\alpha, t) = C_0 \exp(2\Gamma \epsilon_M^2 t)^{1/2} \exp[-2\Gamma(\epsilon_\alpha - \epsilon_M)^2 t], \tag{13}$$

where $\epsilon_M = -Q(t)/2t$ represents the position of the maximum of $C(\epsilon_\alpha, t)$ and changes with time. Employing a saddle-point estimate of the integrals, we obtain from eqn. (5) the following approximation for the structure function:

$$C(\epsilon_\alpha, t) \sim C_0[t^{d_s/4}(\ln t)^{(2-d_s)/4}]^{1-(1-x)^2}, \tag{14}$$

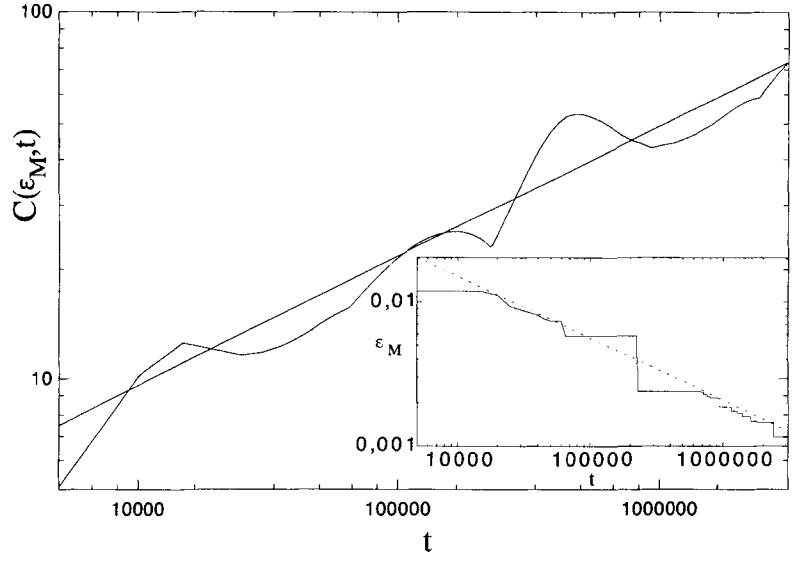
where $x = \epsilon_\alpha/\epsilon_M$. The peaks ϵ_M evolves in time as

$$\epsilon_M(t) = \left(\frac{d_s}{8\Gamma} \frac{\ln t + [(2-d_s)/d_s] \ln(\ln t)}{t}\right)^{1/2}. \tag{15}$$

Since eqn. (15) shows that each mode evolves with its own exponent, the COP dynamics are characterized by multiscaling. More interestingly we observe from fig. 3 that the height of the peak grows with the exponent $d_s/4$ on the average but displays large deviations from a pure power law, reflecting the existence of singularities in the density of states at all energy scales, a feature not observed for regular lattices.

All the results we have discussed are in total agreement with a renormalization group (Marini Bettolo Marconi and Petri 1997a,b) analysis which we have carried out.

Fig. 3



Height of the peak of the structure function for the spherical model and COP case for the $d = 2$ SG. In the inset we show the peak position as a function of time.

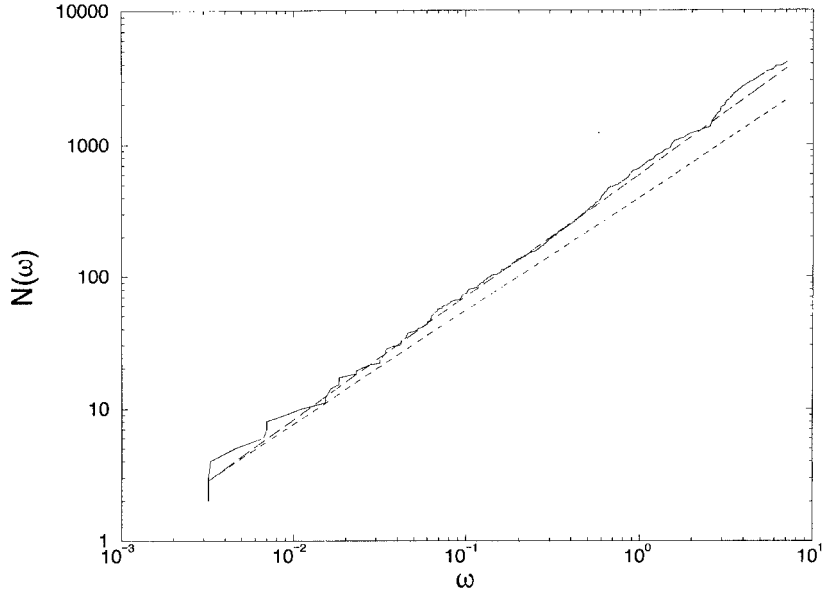
On the other hand we do not expect to observe in real systems properties related to the deviations of the density of states from a smooth behaviour, because the latter are characterized by a less singular density of states than deterministic fractals whose spectrum contains a high degree of correlation which manifests itself through strong correlations between different scales.

§4. Sierpinski carpets

After having seen non-trivial examples of fractals with finite ramification order (i.e. such that after eliminating a finite number of connections one can remove an arbitrary large number of sites from the structure), namely the SG, that admit the exact evaluation of the spectral dimension, we turn out attention infinitely ramified fractals, for which the exact d_s is not known. Even in this case, the spherical model analysed in the previous section can be solved seminumerically on the two-dimensional Sierpinski carpet after, obtaining the spectrum by diagonalization techniques. The fractal dimension is $d_f = \ln 8/\ln 3 = 1{\cdot}91$, while we estimated numerically the fractal dimension to be $d_s = 1{\cdot}86$. This value should be compared with the approximate estimate by Hattori *et al.*† (1985), who give the value $d_s = 2\ln 8/\ln(56/5) = 1{\cdot}721$ slightly smaller than the Migdal–Kadanoff estimate by Watanabe (1985) which is $d_s = 2\ln 8/\ln(56/6) = 1{\cdot}862$. The cumulative density of states is shown in fig. 4. The system does not display a phase transition at any finite value of T_f, since $d_s < 2$. In fig. 5 we display the increase in the peak of the structure function as a function of time in the case of NCOP dynamics for the spherical model

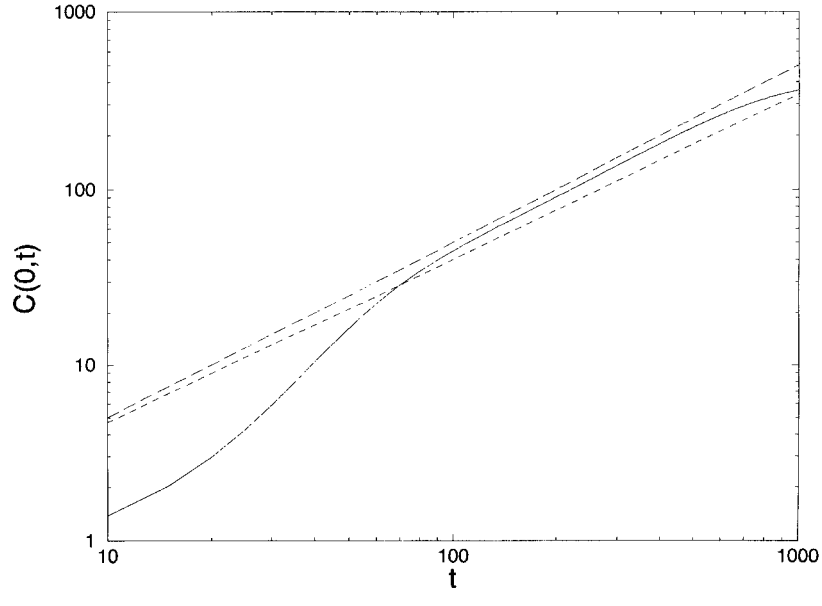
† The equilibrium properties of interfaces in the mode Hartree approximation were studied in this paper.

Fig. 4



Cumulative density of states for a two-dimensional Sierpinski carpet of linear dimension 243. The lines show respectively the slope $d_s = 1$ and $d_s = 0{\cdot}91$.

Fig. 5



Height of the peak of the structure function for the spherical model in the case of a two-dimensional Sierpinski carpet of linear dimension 243.

on a N^2 lattice, with $N = 81$. The logarithmic plot in this case indicates that the growth exponent is between the value corresponding to $d = 2$ and that corresponding to d_s, but we cannot resolve between the two cases with certainty owing to the finite size effects.

§ 5. Dynamical properties of the scalar model on deterministic fractals and stochastic lattices

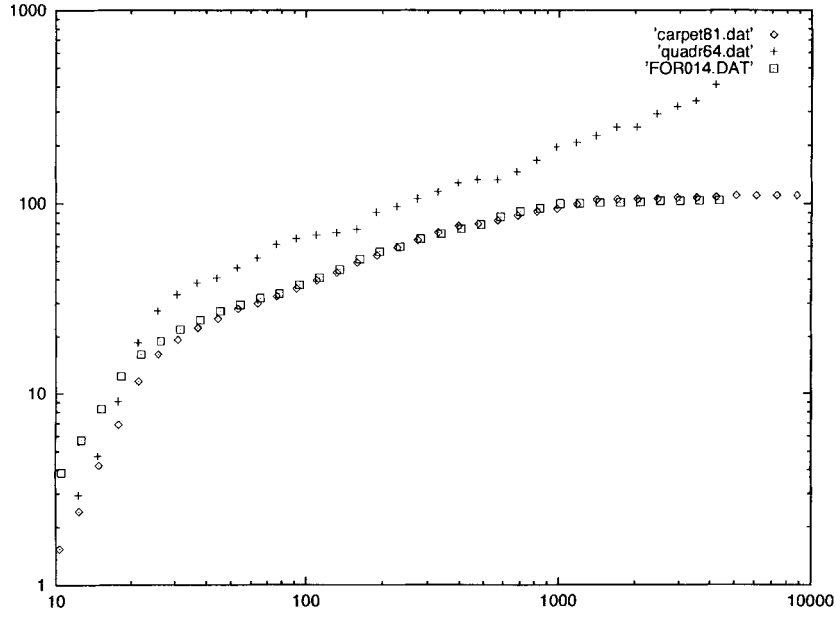
Our findings of the previous sections may represent a starting point to understanding the phase separation occurring in complex disordered structures but refer to a kind of model which is too idealized to be applicable to describing he real-world phase separation. Thus we go beyond the analytical solvable model and consider the case with local nonlinearity on percolation lattices. On the Sierpinski carpet we have analysed the growth of the domains of the minority phase after a quench from an initial inhomogeneous state. To characterize the growth we have measured the number of sites belonging to each domain and the radius of gyration of each domain. The latter was defined as the average

$$R^2 = \frac{1}{M}\sum_{i}^{M}[(x_i - x_{cm})^2 + (y_i - y_{cm})^2].$$

The sum is over the lattice sites of the droplet, (x_i, y_i) is the position of the lattice sites i and (x_{cm}, y_{cm}) the position of its centre of mass.

In fig. 6 we display some preliminary calculations of the growth law for the size and radius of gyration of the droplets of the minority phase of a system with COP

Fig. 6



Domain size against time for the scalar model on a two-dimensional Sierpinski carpet of linear dimension 81.

dynamics. Comparing the data for the average radius relative to the Sierpinski carpet with those for a Euclidean square lattice of the same size we observe that the growth seems to become slower for the first system than for the second. We also observe that during the evolution the interfaces between opposite phases become pinned on bottlenecks separating two blobs. This particular configuration minimizes the interfacial free energy. On the other hand, in the case of non-diluted systems, planar interfaces can be shifted rigidly without free-energy cost, while in the present case an interface localized in a bottleneck corresponds to a local minimum of free energy. Thus the further growth of a domain can take place only through an activation process.

We have indications that after an initial transient the scenario becomes similar to that relative to the Sierpinski carpet.

§6. Conclusions

To summarize, we have obtained growth laws characterized by a new set of non-integer exponents for an exactly solvable model with a vector order parameter.

For the NCOP we confirm the dynamic scaling hypothesis and derive explicitly the scaling form for the structure functions (see eqn. (11)), whereas for the COP we generalize the multiscaling concept.

We have also shown that even on fractal substrates the structure functions display data collapse and thus during the growth there exists dynamical scaling. Of course lattice anisotropies can render the correlation functions orientation dependent and not translationally invariant. On the other hand, the growth exponents at least for the spherical models depend on the substrate through the spectral dimension. At the moment we cannot make quantitative predictions about the scalar model.

We finally remark that perhaps the most serious flaw of the spherical model is the problem of the absence of a region of metastability between the coexistence line and the spinodal line (Binder 1991); the globally constrained model is always unstable inside the two-phase coexistence line. As a consequence, no nucleation barrier needs to be overcome in the transformation from liquid to solid. In the initial state, long-wavelength fluctuations grow and the system becomes unstable. The nucleation barrier is proportional to the surface tension associated with the creation of a kink in the scalar model, whereas in the model with global couplings the energy gaps between the ordered phase and the instanton solutions, that is the uniform solutions of the equation, vanishes in the infinite-volume limit. Further work to clarify the growth regime on these structures is under way.

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